SYNTHESIS OF FULLERENIC MATERIALS BY CONTROLLED PREMIXED COMBUSTION

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Nanostructured carbonaceous materials such as fullerenes and carbon nanotubes are expected to play an increasingly important role in a large range of products including photovoltaic and electronic devices, cosmetics, polymers and biocompatible materials. However, development of the commercial potential of these materials requires their availability at a competitive price. Inexpensive manufacture of high quality C_{60}, C_{70}, higher fullerenes, fullerene black and carbon nanotubes has been achieved by controlled premixed combustion of hydrocarbons. Initially mostly motivated by the need of pollution control of combustion processes, detailed characterization of materials generated at well-defined conditions has shown the potential of combustion technology for the targeted large-scale production of different classes of fullerenic materials.

Conversion of aromatic hydrocarbons to spheroidal fullerenes has been demonstrated at MIT in low-pressure premixed benzene-oxygen flames. Combined yields of C_{60} and C_{70} but without accounting for higher fullerenes, amounting to 20% of the soot produced and 0.5% of the carbon fed, were observed in an early parametric study. After extraction with toluene, high-pressure liquid chromatography (HPLC) using UV-vis and mass spectrometric detection, allowed for the identification of fullerenic compounds up to C_{116}. UV-vis spectra were measured up to C_{108} and isomers could be discerned for C_{78}, C_{90} and C_{94}. Abundances of nearly all fullerenes larger than C_{70} were found to be significantly higher in combustion-generated material than in that produced by electric arc discharge, the initial method for fullerene synthesis. Major improvements for the fullerene production technology were achieved by an increasingly detailed understanding of the chemical processes involved in the formation of different carbonaceous materials such as soot, fullerenes and the competition between both. Optimized operating parameters and process control have led to significantly improved conversion efficiencies of the initial fuel and abundances of extractable spheroidal fullerenes of more than 90%. Assisted by improved purification technology, high-purity fullerenes up to C_{96} are supplied by Nano-C.

Operating a reactor previously used for fullerene synthesis, addition of suitable catalyst precursors to non-sooting hydrocarbon flames leads to the synthesis of single-walled carbon nanotubes (SWCNT). In the present work, iron pentacarbonyl, a liquid with a relatively high vapor pressure, is added to the premixed mixture via an argon stream. The argon flow rate and the temperature in the iron pentacarbonyl reservoir allow controlling its concentration in the fresh gas mixture. Nanotube material has been characterized by Raman spectroscopy, scanning-electron microscopy (SEM), transmission electron microscopy (TEM) and thermogravimetric analysis (TGA). Fuel-rich, non-sooting premixed methane combustion has been found to date to achieve maximized yields of SWCNT with little or no contamination by other carbonaceous materials. SEM and TEM images of materials collected from such flame are shown in Fig. 1. Correlations of the nanotube formation efficiency and the parameters of the combustion process such as the composition of the fresh gas mixture including the initial iron pentacarbonyl concentration will be described. Pathways of iron pentacarbonyl decomposition and subsequent growth of particles catalyzing nanotube nucleation and growth will be discussed.

a) **b)**

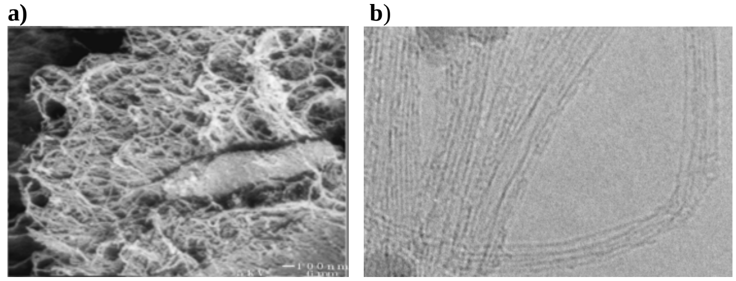

Figure 1. a) SEM and b)TEM of as-produced material.